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Mercuric pollution of surface water, superficial sediments, Nile tilapia (*Oreochromis nilotica* Linnaeus 1758 [Cichlidae]) and yams (*Dioscorea alata*) in auriferous areas of Namukombe stream, Syanyonja, Busia- Uganda

Timothy Omara [Corresp., Equal first author, 1, 2, 3], **Shakilah Karungi** [Equal first author, 4], **Raymond Kalukusu** [2, 5], **Brenda Victoria Nakabuye** [5, 6], **Sarah Kagoya** [2, 7], **Bashir Muasu** [2, 5]

[1] Department of Chemical Engineering, Faculty of Engineering and Informatics, University of Bradford, Bradford, West Yorkshire, BD7 1DP, United Kingdom

[2] Department of Chemistry, Faculty of Science, Kyambogo University, Kampala, Uganda

[3] Department of Quality Control and Quality Assurance, Product Development Directory, AgroWays Uganda Limited, Jinja, Uganda

[4] Department of Mining and Water Resources Engineering, Faculty of Engineering, Busitema University, Tororo, Busitema, Uganda

[5] Department of Quality Control and Quality Assurance, Leading Distillers Uganda Limited, Kampala, Uganda

[6] Department of Food Processing Technology, Faculty of Science, Kyambogo University, Kampala, Uganda

[7] Department of Quality Control and Quality Assurance, Product Development Directory, Sweets and Confectionaries Section, Kakira Sugar Limited, Jinja, Uganda

Corresponding Author: Timothy Omara
Email address: prof.timo2018@gmail.com

The mercuric content, pollution and contamination characteristics of water, sediments, edible muscles of a non-piscivorous fish (*Oreochromis nilotica* Linnaeus 1758 [Cichlidae]) and yams (*Dioscorea alata*) in mercury-based artisanal and small-scale gold mining (ASGM) impacted Namukombe stream and its propinquity, Busia gold district, Uganda were evaluated. Human health risk assessment from consumption of the fishes and yams as well as dermal contact with sediments from the stream were performed. Forty-eight (48) samples of water (12), sediments (12), fish (12), and yams (12) were taken at intervals of 0, 10, 20 and 30m from up, middle and down sluices of the stream and analyzed for total mercury (THg) using US EPA method 1631. Results showed that water in the stream is polluted with mercury (Hg) in the range of 0.00 to 1.21±0.070mg/L while sediments contain Hg up to 0.14±0.04$u$gg^{-1}. THg content of the edible muscles of *Oreochromis nilotica* ranges from 0.00 to 0.11±0.010$u$gg^{-1} while yams contain 0.00 to 0.30±0.001$u$gg^{-1} of Hg. The estimated daily intakes (EDIs) ranged from 0.0049 to 0.0183$u$gg^{-1}day^{-1} and 0.020 to 0.073$u$gg^{-1}day^{-1} for fish consumed by adults and children respectively. The corresponding health risk indices (HRIs) ranged from 0.0123 to 0.04576 and 0.05 to 0.183. EDIs were from 0.0042 to 0.1279$u$gg^{-1}day^{-1} and 0.013 to 0.394$u$gg^{-1}day^{-1}

for yams consumed by adults and children respectively. The HRIs recorded were from 0.011 to 0.320 and to 0.033 to 0.985. All the mean THg contents of the investigated matrices were within acceptable WHO/US EPA limits except for water samples. Consumption of yams grown at 0m up sluice of Namukombe stream may pose deleterious health risks as reflected by the HRI of 0.985 being very close to 1.0. From pollution and risk assessments, Hg usage should be delimited in Syanyonja ASGM areas; solutions to abolish mercury based ASGM in the area ought to be sought at its soonest to avert the accentuating health, economic and ecological disaster arising from the continual discharge of Hg into the surrounding areas. Other safe gold recovery methods such as use of borax should be encouraged. Waste management system for contaminated wastewater, used Hg bottles and tailings should be centralized to enable Hg waste management in ASGM areas in Syanyonja.

Mercuric pollution of surface water, superficial sediments, Nile tilapia (*Oreochromis nilotica* Linnaeus 1758 [Cichlidae]) and yams (*Dioscorea alata*) in auriferous areas of Namukombe stream, Syanyonja, Busia- Uganda

Timothy Omara[1,2,3], Shakilah Karungi[4], Raymond Kalukusu[2,5], Brenda Victoria Nakabuye[5,6] , Sarah Kagoya[2,7], Bashir Musau[2,5]

[1] Department of Chemical Engineering, Faculty of Engineering and Informatics, University of Bradford, West Yorkshire, BD7 1DP, United Kingdom

[2] Department of Chemistry, Faculty of Science, Kyambogo University, Kampala, 1, Kyambogo, Uganda

[3] Department of Quality Control and Quality Assurance, Product Development Directory, AgroWays Uganda Limited, 1924, Jinja, 34-60-Kyabazinga way, Uganda

[4] Department of Mining and Water Resources Engineering, Faculty of Engineering, Busitema University, Tororo, Busitema, Uganda

[5] Department of Quality Control and Quality Assurance, Leading Distillers Uganda Limited, Kampala, 3882/83-Buloba, Wakiso, Uganda

[6] Department of Food Processing Technology, Faculty of Science, Kyambogo University, Kampala, 1, Kyambogo, Uganda

[7] Department of Quality Control and Quality Assurance, Product Development Directory, Sweets and Confectionaries Section, Kakira Sugar Limited, 121-Jinja, Kakira, Uganda

Corresponding Author:
Timothy Omara[1,2,3]
Richmond Road, Bradford, West Yorkshire, BD7 1DP, +447, United Kingdom
Email address: prof.timo2018@gmail.com

Abstract

The mercuric content, pollution and contamination characteristics of water, sediments, edible muscles of a non-piscivorous fish (*Oreochromis nilotica* Linnaeus 1758 [Cichlidae]) and yams (*Dioscorea alata*) in mercury-based artisanal and small-scale gold mining (ASGM) impacted Namukombe stream and its propinquity, Busia gold district, Uganda were evaluated. Human health risk assessment from consumption of the fishes and yams as well as dermal contact with sediments from the stream were performed. Forty-eight (48) samples of water (12), sediments (12), fish (12), and yams (12) were taken at intervals of 0, 10, 20 and 30m from up, middle and down sluices of the stream and analyzed for total mercury (THg) using US EPA method 1631. Results showed that water in the stream is polluted with mercury (Hg) in the range of 0.00 to

1.21±0.070mg/L while sediments contain Hg up to 0.14±0.040μgg^{-1}. THg content of the edible muscles of *Oreochromis nilotica* ranges from 0.00 to 0.11±0.010μgg^{-1} while yams contain 0.00 to 0.30±0.001μgg^{-1} of Hg. The estimated daily intakes (EDIs) ranged from 0.0049 to 0.0183$\mu gg^{-1}day^{-1}$ and 0.020 to 0.073$\mu gg^{-1}day^{-1}$ for fish consumed by adults and children respectively. The corresponding health risk indices (HRIs) ranged from 0.0123 to 0.04576 and 0.05 to 0.183. EDIs were from 0.0042 to 0.1279$\mu gg^{-1}day^{-1}$ and 0.013 to 0.394$\mu gg^{-1}day^{-1}$ for yams consumed by adults and children respectively. The HRIs recorded were from 0.011 to 0.320 and to 0.033 to 0.985. All the mean THg contents of the investigated matrices were within acceptable WHO/US EPA limits except for water samples. Consumption of yams grown at 0m up sluice of Namukombe stream may pose deleterious health risks as reflected by the HRI of 0.985 being very close to 1.0. From pollution and risk assessments, Hg usage should be delimited in Syanyonja ASGM areas; solutions to abolish mercury based ASGM in the area ought to be sought at its soonest to avert the accentuating health, economic and ecological disaster arising from the continual discharge of Hg into the surrounding areas. Other safe gold recovery methods such as use of borax should be encouraged. Waste management system for contaminated wastewater, used Hg bottles and tailings should be centralized to enable Hg waste management in ASGM areas in Syanyonja.

Introduction

Artisanal and small-scale mining (ASM) is a lucrative source of income in Uganda employing over 400,000 people as of 2015 [1]. ASM in Uganda focuses majorly on gold in the gold districts of Mubende, Namayingo, Busia, Buhweju, Kaabong, Nakapiripirit and Amudat; sand (countrywide), clay, tin, wolfram and iron ore in Isingiro, Ntungamo, Kabale, Kisoro and Kanungu districts. The discovery of gold (Au) was first witnessed in West Nile in 1915, but no mining commenced until 1933. In Busia, gold was reported in 1932 in Osipiri area (Busia-Kakamega greenstone belt) [2] which registered ASGM on vein and alluvial deposits in the auriferous areas of Tiira, Makina, Amonikakine and Osapiri villages up-to-date [3]. Despite the fact that rudimentary tools are employed, ASGM in Busia has continued, an indication that the business is recovering sizeable quantities of gold nuggets [3]. More so, gold prices now exceeds US $1,600 per ounce (from less than US$500 in the 1980s), causing ASGM to rise along with its elemental mercuric pollution [4]. Reports from Bank of Uganda noted gold as the second most important export of Uganda after coffee with a worth of US $35.73m in 2015 and US $339.54m in 2016 [5].

In Busia and Bugiri districts, close to 1,000 gold miners engage in mercurial ASGM with reported 150kg of Hg per annum ending up in the downstream areas. Approximated 45kg of Hg per annum get discharged with tailings into small rivers and streams in Busia during gravity concentration of auriferous materials (panning) [6]. Miners are thus exposed to elemental Hg intoxication [7] as well as cyanide used to extract gold from tailings. Similarly, miners handling amalgamation Hg or living in close proximity to the uncontrolled mercurial gold recovering sites risk coming in contact with Hg through dermal adsorption during amalgamation, inhalation of

Hg vapor, drinking Hg polluted water and consumption of foods such as fish, yams, maize, millet and potatoes grown in Hg-contaminated water and soils. The ASGM population of the neighboring village to Syanyonja (Tiira) who participated in mercurial ASGM have been diagnosed with severe health complications including paralysis [8]. Worse still, many miners have been buried by open pits in mines. The miners reportedly use bare hands during gold recovery without usage of personal protective equipment [8] contrary to international guidelines (UNIDO) on Hg use [1]. It is reported that Hg is poorly recovered in ASGM processes and the emission factor can be as high as 15g Hg/g gold produced [9]. More so, ASGM is inextricably linked with human health and poverty concerns.

Elemental Hg intoxication leads to irreversible neurological, kidney and autoimmune impairment coupled with respiratory tract irritation, chemical pneumonitis, pulmonary oedema, chest tightness, respiratory distress [10], respiratory failure and death [11]. Systemic absorption of elemental Hg via the lungs causes nausea, vomiting, headache, fever, chills, abdominal cramps, and diarrhea. Chronic, lower level exposure to elemental Hg induces gingivostomatitis, photophobia, tremors and neuropsychiatric symptoms (fatigue, insomnia, anorexia, shyness, withdrawal, depression, nervousness, irritability and memory problems) [12]. Elemental and inorganic Hg toxicity in children may be witnessed in oedematous, painful, red, desquamating fingers and toes (acrodynia), as well as hypertension [13].

Mercury-gold amalgamation transmogrifies elemental Hg into methylmercury (MeHg), which is the most toxic organic form of Hg and a powerful neurotoxin reported to enter food chain through bioaccumulation [13,14]. MeHg because of its lipid-solubility can readily enter bloodstream via the digestive system, usually in excess of 90% [15]. On crossing the blood-brain barrier, MeHg accumulates in the spinal cord, triggering headaches, ataxia, dysarthria, visual field constriction, blindness, hearing impairment, psychiatric disturbance, muscle tremor, movement disorders, paralysis and death [16].

Uganda National Environmental Management Authority (NEMA) reported in 2017 that the Ugandan ASGM sector contributes an estimated annual Hg input of 18.495Mt per annum, of which 12.136 Mt per annum is released in the air, 3.333Mt per annum is released in water, and 3.027Mt per annum is disposed on land [17]**.** As reported in other ASGM areas globally, Hg from ASGM is often discharged in a perverted fashion into ecosystems [18,19], initiating prodigious pollution of aquatic biota, water, sediments, soil and air [20-22]. This is disastrous for the case of Syanyonja and Busia as a whole since the Hg may likely get entrained in sediments which can enter ‘‘the life artery of East African countries’’ (Lake Victoria), 30km downstream via a series of wetlands [6].

This study provides the first ever comprehensive assessment of mercuric contamination of water, sediments, fish and yams from Namukombe stream in Syanyonja village, Busia district and creates a paradigm for future studies on the development of effective remediation strategies on reducing mercuric pollution from ASGM in Busia and could improve government decision-making on ASGM activities in the gold districts of Uganda.

Materials and Methods

Brief description of the area under study

The study was conducted in Namukombe stream, Syanyonja village, Busitema sub county, Busia gold district, Eastern Uganda (**Figure 1**). Syanyonja village lies in the coordinates UTM 60648 36N617118. Namukombe stream is the one of the water bodies in Syanyonja village where mercury-based ASGM activities, washing, bathing, fishing and growing of food crops are done.

Collection and preparation of samples

All samples were obtained in triplicate from up, middle and downstream at 0,10, 20 and 30m along the stream.

Water samples were collected in plastic containers precleaned by washing in non-ionic detergent, rinsed with tap water and later soaked in 10% HNO_3 for 72hours and finally rinsed with deionized water prior to usage. The collected samples were microfiltered through 0.45µm pore diameter membrane filters immediately and preserved with concentrated nitric acid. For analysis, the samples were prepared using the American Public Health Association (APHA) standard method 9221 for examination of water and wastewater [23].

Superficial sediment samples were obtained using a grab sampler at 0-5cm in accordance with the United Nations Environment Programme's reference method for sediment pollution studies (UNEP method number RSRM 20) [24] to address geographic differences. Sediment samples collected were put in clean presterilized plastic bottles, tightly sealed and labelled. The samples were oven-dried at 80-95°C for 16 hours to reach a constant weight, then crushed in a stone mortar and sieved through a 63µm sieve. The powdered samples were preserved at 4^0C on an ice block to preserve their integrity prior to analysis.

The best way to determine if mercury is becoming bioavailable within discrete areas is to analyze resident biota, such as invertebrates or small fish. Fish samples (5.5-8.0cm fork length) were caught and preliminarily prepared for analysis as described by Omara *et al.* [25].

Yam samples obtained were peeled and sliced. Aliquots (3.0±0.1g) were weighed and ashed in a furnace at 550^{o}C for 5 hours.

Analysis of samples

Total mercury in the samples were determined as per the US EPA method 1631 [26]. Total mercury was analyzed by BrCl oxidation and immediately prior to analysis, excess bromine in all samples was neutralized with 10% hydroxylamine hydrochloride. All samples were then reduced with stannous chloride, purged with nitrogen gas and trapped on columns packed with gold coated sand. The gold trap was heated and the desorped mercury detected with Cold-Vapor Atomic Fluorescence Spectrophotometer- CVAFS. The concentrations were reported in mg/L for water or $\mu g g^{-1}$ for solid foods for easy comparison with set international compliance limits.

Human health risk assessment

The estimated daily intake (EDI) was calculated to averagely estimate the daily Hg loading into the body system of a specified body weight of a consumer (adult/child) via consumption of contaminated fish and yams while the average daily dose (ADD_{therm}) was calculated to determine intoxication via dermal contact with mercurially polluted sediments. These provide the relative availability of Hg but does not take into cognizance the possibility of metabolic ejection of Hg. EDI in $\mu g g^{-1} day^{-1}$ was computed using **Equation 1** previously employed elsewhere [25] while ADD_{therm} ($\mu g g^{-1} day^{-1}$) was computed from **Equation 2** [27-29].

$$EDI = \frac{E_f \times E_d \times F_{ir} \times C_f \times C_{hm}}{W_{ab} \times T_{aet}} \quad (1)$$

$$ADD_{therm} = \frac{C_{hm} \times S_A \times AF \times E_f \times E_d}{W_{ab} \times T_{aet}} \times 10^{-6} \quad (2)$$

Where E_f = exposure frequency (365 days/year); E_d = exposure duration, the average lifetime (58.65 years for an adult Ugandan) [30]; F_{ir} is the fresh food ingestion rate (g/person/day) = 48 for fish and 301.0 and 231.5 for yams for adults and children respectively) [31,32]; C_f is the conversion factor (0.208) for fresh weight (F_w) to dry weight (D_w) for fish and 0.085 (to convert fresh yams weight to dry weight; considering it as a vegetable) [33], C_{hm} = heavy metal concentration in foodstuffs ($\mu g g^{-1}$ *Fw*); W_{ab} = average body weight (considered to be 15kg for children [29] and 60 kg for adults [34]) and T_{aet} = average exposure time for non-carcinogens (given by the product of E_d and E_f) [35]; S_A , the exposed surface area in cm^2 = 4,350 for adults and 2,800 for children [29]; AF is the skin adherence factor in $mg/cm^2/day$ = 0.7 for adults [36] and 0.2 for children [29].

Health Risk Index (HRI), the total risk of non-carcinogenic element via three exposure pathways was evaluated using Target Hazard Quotient (THQ) in accordance with US EPA Region III risk-based concentration table [37] used in a preceding study [25]. According to the criterion, THQ (HRI) less than unity (1.0) implies the exposure is very unlikely to have adverse effects whereas THQ greater than unity prognosticates a possibility of non-carcinogenic effects, with an increasing probability as the value increases [31]. The numerical HRI i.e. THQ values were obtained from the ratio of EDI or ADD_{therm} to the Reference Dose (R_fD) **(Equation 3)** [25, 28].

$$THQ = \frac{EDI}{R_fD} \quad \text{or} \quad THQ = \frac{ADD_{therm}}{R_fD} \quad (3)$$

The R_fD of Hg via ingestion in µg/g/day is 4.0×10^{-1} [37] while the R_fD for Hg via dermal contact is 1.0×10^{-2} µg/g/day [29].

The R_fD (usually in mg/kg/day) is the maximum daily dose of a metal from a specific exposure pathway, that is believed not to lead to an appreciable risk of deleterious effects to sensitive individuals during a life time [38]. If the EDI is lower than the R_fD, the THQ is less than 1, and adverse health effects are unlikely to appear, whereas if the EDI exceeds the R_fD, THQ > 1, adverse health effects are likely to appear [27, 28]. In this study, the THQ was calculated basing

on three pathways i.e. consumption of mercurially contaminated fish and yams and dermal contact (assuming miners are occupationally in direct contact with dredged Hg-contaminated sediments). The assumptions made during the health risk calculations were that the ingested dose is equal to the absorbed trace metal dose [39] and that cooking of the fishes and yams have no appreciable effect on the Hg content of the assessed matrices [40].

Assessment of bioaccumulation factors

Bioaccumulation factors (BAFs) are multipliers often employed to estimate concentrations of chemicals that accumulate in tissues through any route of exposure [41]. They are often referred to as bioconcentration factors (BCFs) for aquatic invertebrates. The BCFs and biota to sediment accumulation factor (BSAF) of trace metals from sediment or surface water to animal tissues can be determined for different samples. Thus, BCF was determined from the numerical ratio of concentration of the priority trace metal (Hg) in the whole edible tissues of Nile tilapia to that of concentration of Hg in water while BSAF was determined from the ratio of the concentration of Hg in the edible tissues of *Oreochromis nilotica* to that of Hg in the corresponding sediment samples [42].

Sediment quality assessment

To assess the level of contamination, the environmental and health risks that originate from a heavy metal's occurrence, indices are used to indicate the enrichment of a given environmental component as compared to its natural concentration. The indices used to describe heavy metal enrichment of sediments include contamination factor (CF), enrichment factor (EF), pollution load index (PLI), geoaccumulation index (I_{geo}), potential ecological risk (RI) and hazard quotient (HQ) [43-46].

Single indices of the above-mentioned list are used, as a rule at thumb, to subtly assess sediment contamination. This approach, though allows evaluation of contamination, limits the ability to compare degree of contamination of sediments investigated in different studies. In order to identify pollution problems, the anthropogenic contributions should be distinguished exclusively from the natural sources. Thus, the degree of mercuric pollution of sediments from Namukombe stream was assessed using contamination factor and geoaccumulation index.

The contamination factor (CF) was calculated using **Equation 4** given by Hakanson [43]**.**

$$CF = \frac{C_{hm}}{C_b} \qquad (4)$$

where C_{hm} is the priority trace metal concentration in the analyzed sample and C_b is the geochemical background trace metal concentration/preindustrial concentration. A background concentration of $0.25 \mu g g^{-1}$ was used in this study.

Geoaccumulation index, Igeo for the sediments from the different sluices were obtained from computations utilizing **Equation 5** suggested by Müller [47].

$$I_{geo} = \mathrm{Log}_2 \left(\frac{C_n}{1.5B_n}\right) \qquad (5)$$

From which C_n is the concentration of the trace metal (n) in the sampled and analyzed sediment, B_n is the background concentration of the same metal (n) and the factor 1.5 is the background matrix correction factor due to lithogenic effects (takes into account possible lithological variability) [48,49].

Results

Statistical analysis of results

Analytical data was subjected to statistical evaluation using GraphPad Prism (v7.05.237, GraphPad software, California, USA). Analytical results, unless otherwise specified, were presented as mean ± standard deviations of triplicate analyses. Independent sample test or one-way analysis of variance (ANOVA) followed by LSD post hoc test was used for comparison of the differences between the experimental mean THg concentrations and WHO/US EPA maximum permissible limits. These statistical analyses were performed at a 95% confidence interval (with differences in mean values accepted as being significant at $p < 0.05$).

Mercuric pollution of Namukombe stream

The metalliferous content of water, sediments, fish and yams in the different sluices of Namukombe stream with their descriptive statistics are given in **Table 1**.

Human health risk assessments

The toxicity indices from consumption of fish, yams and dermal contact with sediments from Namukombe stream are given in **Table 2**, **Table 3** and **Table 4**.

Bioaccumulation factors

The statistical bioaccumulation factors computed are given in the **Table 5**.

Sediment quality

The contamination factor and the Müller geoaccumulation index for the sediments from the different sluices are presented in **Table 6.**

Discussion

Mercuric pollution of surface water from Namukombe stream

Total mercurial content of the water samples ranged from 0.00±0.00 to 1.21±0.070mg/L. Hg levels upstream were initially high at 0 m but reduced significantly ($p = 0.0001$) after a distance of 10 m from the point source of ASGM. There was a gradual decrease from 1.21±0.070 to 0.09±0.006mg/L upstream, 0.18±0.010 to 0.02±0.001mg/L in middle stream and 0.10±0.001mg/L until no detection downstream (**Table 1)**.

The observed decrement could have been due to the fact that Namukombe stream is swampy, thus the flow of water is reasonably slow. Since the stream contains organic matter, most of the Hg could have been retained within the sediments at upsluice and mid sluice, making them undetectable downstream. It is reported that organic matter can increase Hg methylation by stimulating heterotrophic bacteria (*Pseudomonas* species) in aerobic conditions [50] or abiotic methylation [51]. In anaerobic condition, Hg reacts with organic carbon in the sediments to form toxic methyl and di-methyl Hg [51]. Some anaerobic bacteria that possess methane synthetase are also reported to be capable of Hg methylation [52]. Once MeHg is released from microbes, it enters the food-chain as a consequence of rapid diffusion and tight binding to proteins in aquatic biota.

Further, owing to their static nature, sediments tend to get enriched with toxic materials than water, which can undergo relatively rapid self-purification. Thus, a greater percentage of THg in an aquatic system is expected in sediments if there is effective binding with organic carbon bearing particles. This may innocuously retard the transfer of Hg to overlying water through interstitial water [53]. Heavy metals which are less soluble in water such as Hg are easily adsorbed and accumulated in sediments [54]; however, in the event that the trace metal cannot be permanently adsorbed by sediments, it is released back to the overlying water, when environmental conditions such as salinity, resuspension, pH, redox potential and the organic matter decay rate changes [55,56]. Thus, this explain the high levels of THg recorded in water than sediments for the corresponding sluices.

The results of this study is comparable (though higher) to that of Oladipo *et al.* [57] where the the mean THg content of water in ASGM areas of Manyera river, Nigeria was recorded at 0.021±0.004mg/L. Mahre *et al.* [58] reported that water from River Kaduna, Nigeria had THg ranging from 1.72 to 2.50mg/L which is quite higher than the maximum mean THg level (1.21±0.070mg/L) recorded in this study. The results from this study agrees well with preceding investigations which concluded that the quality of water in the periphery of Hg-based ASGM sites in Uganda have been innocuously deteriorated by Hg pollution [59,60]. All the THg concentrations of the water samples in this study were higher the US EPA maximum contamination level of 0.002mg/L for Hg in drinking water. Therefore, this water is not safe for drinking and domestic use.

Mercuric content of sediments

Sediments are good hosts of highly toxic pollutants from natural and anthropogenic sources [61] and have been reported as the biggest sink and major reservoir for heavy metals [62-69]. They enhance accumulation of heavy metals in benthic invertebrates, thereby transferring them to higher levels of food chains [70-74]. Therefore, monitoring sediments can enhance a more accurate tracking of trace metal contamination of aquatic ecosystems [75-81] compared to water and/or floating aquatic plants, which tend to give inaccurate estimations due to water discharge fluctuations and lower resident times.

Mean THg concentrations of sediments from up sluice ranged from 0.00±0.00 to 0.14±0.040µgg^{-1}, middle stream ranged from 0.00±0.00 to 0.11±0.050µgg^{-1} while downstream ranged from 0.00±0.00 to 0.12±0.016µgg^{-1} (**Table 1**). All the mean THg concentration in the sediments from the three sluices along the stream is lower than the maximum permissible limit of 0.15µgg^{-1} recommended by USEPA 2001 standard [26]. For all the sluices, THg concentration in the sediments reduced significantly ($p = 0.000994$) from upstream to downstream with the result that no Hg was detected in sediments sampled 30m away from all the point sources.

The mean THg content of the sediments in this study is lower than the values registered in other global studies such as 0.265µgg^{-1} reported by Donkor *et al.* [82] in Pra river (Ghana), 0.7-9.3µgg^{-1} reported by Ramirez-Requelme *et al.* [83] in Amazon and 0.3-0.9µgg^{-1} recorded by Feng *et al.* [84] in Shaanxi Province of the Peoples` Republic of China. However, Lasut *et al.* [85] in Indonesia, Pataranawata *et al.* [86] in Thailand, Mohan *et al.* [51] in Nilambur, Kerala-India and Oladipo *et al* [57] in Manyera river, Nigeria reported lower THg content of sediments of 0.010-0.017, 0.096-0.402, 0.103-0.46, and 0.018µgg^{-1} respectively which are comparable to the mean THg concentrations recorded in this study. Taylor *et al.* [87] reported that the drainage sediments from upstream of Uvinza on the Malagarasi river (Tanzania) contain THg in the range of 0.17–0.24 µg/kg, which were lower than for sediments from Ilagala with 0.10 to 0.66 mg/kg THg but all higher than the mean THg registered in this study.

THg content of all the sediment samples except one i.e. sample from 0m up sluice (0.14±0.040µgg^{-1}) were below Threshold Effect Level (TEL) of 0.13µgg^{-1}. All the THg concentrations of the sediments were lower than the Probable Effect Level (PEL) of 0.70µgg^{-1} for Hg in sediments postulated by Smith *et al*. [88] and MacDonald [89]. THg in the sediments lying between TEL and PEL is expected to be associated with adverse biological effects [51]. Also, among the bottom sediment samples, none had THg higher than the background concentration of 0.25µgg^{-1}, which is considered as normal in non-contaminated sediments [90].

It is worth noting that the mercurial content of the sediments in this stream were lower than the THg content of water from the corresponding sluices. In this study, the retention rates of Hg in sediments, which is influenced by many factors such as the metallic forms of mercury (i.e. elemental, ionic, organic, or inorganic), pH, temperature, organic carbon and electrical conductivity were not investigated. Because the sediment Hg retention rates can vary from one location to another, the observed variability in THg concentrations in sediments from the

different sampled study sites in this study can be attributed to the differences in the sediment Hg retention rates and the level of mercuric pollution due to ASGM activities.
According to the Sediment Quality Criteria for Protection of Aquatic Life (Environment Canada, 1992 cited in [91]), all the sediments had THg below the toxic threshold of 1.0μgg^{-1} and minimal effects threshold of 0.20μgg^{-1}.

Mercuric content of *Oreochromis nilotica* Linnaeus 1758 (Cichlidae)

Fish from all the sluices had mean THg in the range of 0.00±0.00 to 0.11±0.010μgg^{-1} (**Table 1**). All the fishes from the stream did not exceed the maximum WHO permissible limit for Hg in fish for human consumption (0.50μgg^{-1}) as well as the WHO recommended limit for vulnerable groups (0.20μgg^{-1}).
The mean THg content of fish reported in this study is lower than 0.58 ± 0.44μgg^{-1} mean THg reported by Castilhos *et al* [92] in fresh water fish from Tatelu gold mining area, Indonesia which recorded more than 45% of the fishes with THg above WHO compliance limit. Oladipo *et al.* [57] reported that fish (*Heterotis niloticus)* from Manyera river, Nigeria had a THg content of 8.0×10^{-3}μgg^{-1}, well lower than is reported in *Oreochromis nilotica* Linnaeus 1758 (Cichlidae) edible tissues by this study. Mahre *et al.* [58] reported a lower mean THg of 10^{-4} to 10^{-3}μgg^{-1} in fisheries and aquatic life of river Kaduna, Nigeria.
In this investigation, fish samples obtained from upstream close to mining sites had higher THg content than those from downstream (**Table 1**). This could be due to a reduction in the Hg content of water as it flows downstream. Mercury in water could have probably got entrained in the sediments.
It is reported that fish ingest heavy metals by direct uptake in aqueous solution or by epithelial ingestion of trace metal contaminated water that sluices through their gills, skin, oral cavity and digestive tract [93]. However, chronic intake of heavy metals by fish rests entirely on the trace metal concentration, volume of the ingested contaminated food, the heavy metal uptake speed, exposure duration, uptake route, ecological conditions external to the fish (including availability of water, temperature, pH) and innate factors notably fish age [94], fish nutritional habits as well as the dynamic processes involved in the trace metal metabolism [95-97]. Therefore, the lower levels of Hg recorded in this study could be because the fish samples were not so aged and the fact that *Oreochromis nilotica* Linnaeus 1758 (Cichlidae) is non-piscivorous. This is corroborated by the reports of Mol *et al.* [98] who reported that the THg concentrations in freshwater piscivorous fish species in ASGM areas of Suriname, South America was 0.71 μgg^{-1}, well (3.7 times) higher than 0.19μgg^{-1} recorded in non-piscivorous species in the same mercurially contaminated water bodies.
Weber *et al.* [99] pointed that aquatic organisms, including fish, exposed to copious levels of waterborne trace metals bioconcentrate the metals upon absorption, ultimately transferring them to humans as they are inevitable in human nutrition. Thus, for the general population, dietary intake is the dominant exposure pathway to Hg. Extensive investigations have quoted that 75–95% of Hg in most fish exists as MeHg. Therefore, though the levels of THg in the edibles fish

muscles eaten by the residents of Syanyonja registered in this study are evidently low, the effect of its accumulation should not be overruled as other organs of accumulation such as the gills, liver and kidneys might contain higher THg concentrations [25].

More so, chronic exposure to MeHg via consumption of fish and other marine species is a major concern for human health, especially developmental exposure that triggers neurological alterations [100-105]. Hg exposure has been proven to cause elevated risks of cardiovascular diseases with severe exposures causing negative impacts to the reproductive and immune systems [106,107].

Mercuric content of yams (*Dioscorea alata*)

THg content of yams from Namukombe stream varied between 0.00±0.00 to 0.30±0.001μgg^{-1} (**Table 1**). Yams from upstream (at 0m) had the highest mean THg of 0.30±0.001μgg^{-1}. Middle stream samples at 0m had THg content of 0.28±0.014μgg^{-1}, while downstream samples at 0m had the highest mean THg content of 0.29±0.003μgg^{-1} (**Table 1**). There was no significant difference ($p = 0.0004$) in the THg content of the yams from the different sluices. This trend can be related to the levels of Hg in both water and sediments from the sluices in relation to the distance of the samples from the ASGM activities. The highest mercurial content of the yams in Namukombe stream is quite higher than that reported in Rwamagasa ASGM area, Tanzania by Taylor *et al*. [87] where yams recorded THg content of 0.007 to 0.092μgg^{-1}. It is noteworthy that yams in this study recorded the highest THg (0.30±0.001μgg^{-1}) of all the studied matrices. This could be because yams are exposed to the different uptake routes such as the sediments (soils), contaminated water and atmospheric disposition on leaves during growth.

Health risk assessment from consumption of fish and yams and dermal contact with sediments from Namukombe stream

Chronic low level intake of priority trace metals such as Hg have been implicated for deleterious human health effects, which becomes apparent following years of persistent exposure [108-110]. THQ method was used to assess the potential health risks of Hg accumulation through consumption of the edible muscles of fish and yams as well as dermal contact during ASGM.

The estimated daily intakes (EDIs) ranged from 0.0049 to 0.0183μgg^{-1}day^{-1} and 0.020 to 0.073 μgg^{-1}day^{-1} for fish consumed by adults and children respectively. The corresponding health risk indices (HRIs) ranged from 0.0123 to 0.04576 and 0.05 to 0.183 (**Table 2; Table 3**). In 1960s, Minamata residents of Japan suffered unprecedented neuropathies due to the consumption of MeHg-contaminated seafood [111]. More so, some studies have reported that Hg is toxic to fishes (*Tilapia guineersis*, Mugil and *Tilapia fuscatus*) and induces fish weight loss even on exposure to sub-lethal doses in more than two fortnights [112]. Thus, it can be deduced that *Oreochromis nilotica* in Namukombe stream is endangered.

The EDIs were from 0.0042 to 0.1279μgg^{-1}day^{-1} and 0.013 to 0.394μgg^{-1}day^{-1} for yams consumed by adults and children respectively. The statistical HRIs recorded were from 0.011 to 0.320 and to 0.033 to 0.985 respectively (**Table 2; Table 3**). The HRI of 0.985 registered for

consumption of yams from 0m upsluice by children is very close to 1.0, implying that consumption of yams from this site by children might lead to mercury-related health risks.

The ADD_{therm} ranged from 1.015×10^{-6} to 7.105×10^{-6} $\mu gg^{-1}day^{-1}$ and 7.47×10^{-7} to 5.227×10^{-6} $\mu gg^{-1}day^{-1}$ (**Table 4)** for dermal contact with mercury-contaminated dredged sediments from Namukombe stream by adults and children respectively. The HRIs respectively ranged from 1.015×10^{-4} to 7.105×10^{-4} and 7.47×10^{-5} to 5.227×10^{-4} for adults and children (**Table 4)**.

THQ of less than unity (1.0) indicate the relative absence of health risks associated with intake of Hg through consumption of either Hg contaminated fish, yams or dermal contact with sediments. However, ingestion of both fish and yams, coupled with persistent dermal exposure to Hg in sediments during panning would lead to potential health risks especially for children.

Mercuric accumulation based on bioaccumulation factors

The bioaccumulation factors (BAFs), bioconcentration factor (BCF) and biota to sediment accumulation factor (BSAF) computed for *Oreochromis nilotica* Linnaeus 1758 (Cichlidae) in Namukombe stream are presented in **Table 5**. The results show a more significant increase in Hg levels in *Oreochromis nilotica* Linnaeus 1758 (Cichlidae) tissues than in the surface water samples. BCF values for Hg in *Oreochromis nilotica* Linnaeus 1758 (Cichlidae) were ranked as follows: downstream > middle stream > upstream. The highest BCF of **0.800** was recorded at 0m downstream while the lowest BCF of **0.250** was recorded at 10m middle stream. Such trace metal accumulation levels in fish as in this concerted study augment published data reported by other authors on different species of aquatic organisms [42, 113-115]. Therefore, this study suggests that *Oreochromis nilotica* Linnaeus 1758 (Cichlidae) is a sentinel organism for biomonitoring of aquatic ecosystems.

BSAF explored the rate of Hg uptake from the sediment and its subsequent accumulation in *Oreochromis nilotica* Linnaeus 1758 (Cichlidae) tissues. In this investigation, the highest BSAF value of **1.500** was recorded at 10m middle stream while the lowest BSAF (**0.333**) was recorded at 10m upstream. Thus, Hg enrichment was highest in the middle stream, though the sediments have higher concentrations of Hg than the edible muscles of *Oreochromis nilotica* Linnaeus 1758 (Cichlidae).

Quality of superficial sediments from Namukombe stream

Contamination factor

All the statistical CFs were less than 1.0 (the highest statistical value of **0.56** was recorded at 0m upstream and the lowest value of **0.04** was reported at 10m downstream) (**Table 6**). According to Hakanson [43], four (4) contamination categories are distinguished: CF <1: low contamination, $1 \leq CF < 3$: moderate contamination, $3 \leq CF < 6$: considerable contamination and $CF > 6$: very high contamination. Thus, basing on the aforeacknolwedged criteria, there is very low contamination of the sediments of Namukombe stream.

Geoaccumulation Index

Müller geoaccumulation index (I_{geo}) is a frequently employed analytical index for examination of the contamination level of sediment samples by trace metals. It assesses the degree of contamination by comparing the current levels of trace metal concentrations to the previous status of the research site. The computed Müller geoaccumulation indices for the bottom sediments from Namukombe stream ranged from -5.233 to -1.423 **(Table 6)**.

The I_{geo} is composed of seven grades along with associated sediment quality levels according to the degree of trace metal pollution. The values are classified as follows: no contamination (I_{geo}<0); low to median contamination (I_{geo} between 0 and 1); median contamination (I_{geo} between 1 and 2); median to strong contamination (I_{geo} between 2 and 3); serious contamination (I_{geo} between 3 and 4); serious to extreme contamination (I_{geo} between 4 and 5); and extreme contamination (I_{geo}>5).

In this study, the geoaccumulation indices were all negative for the sluices **(Table 6)**, reflecting that there is no serious anthropogenic (mercuric) pollution of the studied sites in Namukombe stream.

Conclusions and recommendations

Persistent utilization of Hg in ASGM in Syanyonja and the proliferation of its environmental and human health effects pose significant challenges to sustainability; water in Namukombe stream is contaminated with up to 1.21±0.070mg/L of Hg which is above US EPA maximum permissible limit for Hg in drinking water. The maximum THg content of sediments from the stream is 0.14±0.040μgg^{-1} which is lower than the maximum limit of 0.150μgg^{-1} recommended by USEPA 2001 standard. Release of ASGM residual Hg into Namukombe stream have resulted in significant entrainment of Hg in water and sediments in the stream. The mercuric content of the edible whole muscles of the locally consumed fish (*Oreochromis nilotica* Linnaeus 1758 [Cichlidae]) is lower than that reported in sediments, yams and drinking water.

THg content of the edible whole muscles of fish from Namukombe stream ranges from 0.00±0.00 to 0.11±0.010μgg^{-1} which is still within the maximum WHO permissible limit of 0.5μgg^{-1} for Hg in fish for human consumption. Health risk assessment indicates that consumption of yams from 0m up sluice may have potential health risk, particularly to children. From pollution assessment, mercury usage should be delimited in Syanyonja ASGM areas; strategies to minimize or abolish mercurial ASGM in the area should be reached to avert the accentuating health, economic and ecological disaster arising from the continual discharge of Hg into the surrounding areas. Other safe gold recovery methods such as use of borax should be encouraged. Waste management system for waste wastewater, used Hg bottles and tailings should be centralized to enable Hg waste management in ASGM areas in Syanyonja.

Further research should determine the geochemical properties (pH, organic carbon and conductivity) of the sediments as these properties tend to correlate with Hg accumulation in sediments. Research should be done to evaluate the mercuric content of the different organs of

accumulation (gills, liver, kidney) of *Oreochromis nilotica* Linnaeus 1758 (Cichlidae). The levels of methyl mercury and other trace metals such as Lead and Arsenic should be determined in water, sediments, yams, fish as well as soils. The atmospheric flux of mercury in the atmosphere of the study area should be determined.

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Figure 1

Map of the area under study

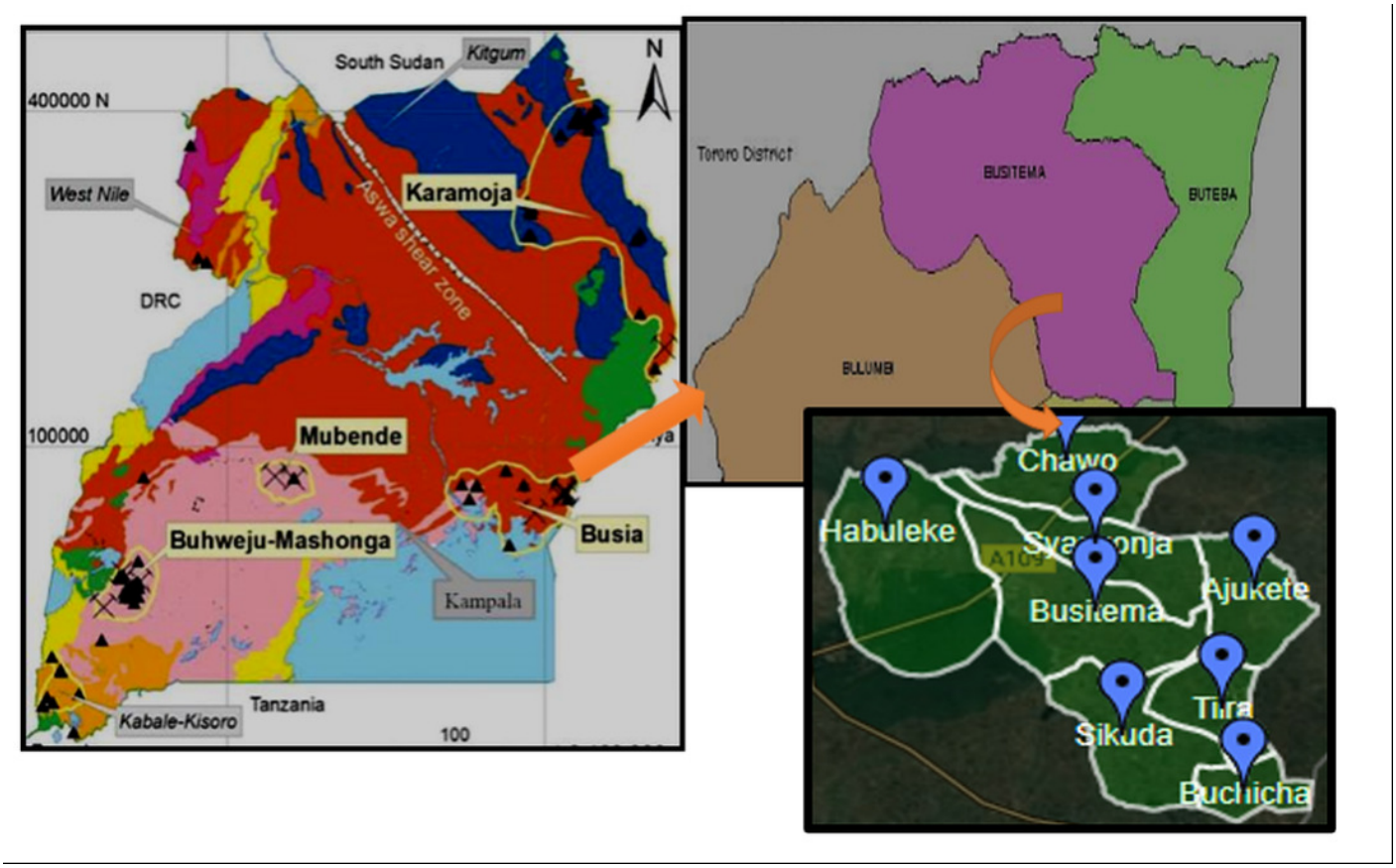

Table 1(on next page)

Mercurial Content of water, sediments, fishes and yams

Table 1. Mercurial content of water, sediments, fish and yams in Namukobe stream

Sample	Distance (m)	Mean total mercury concentration (mg/L or μgg^{-1})											
		Up sluice				Middle sluice				Down sluice			
		Mean ± S.D	Range	S.E	Variance	Mean ± S.D	Range	S.E	Variance	Mean ± S.D	Range	S.E	Variance
Water	0	1.21±0.040	1.17-1.25	0.023	0.0016	0.18±0.070	0.11-0.25	0.041	0.0049	0.10±0.030	0.07-0.13	0.017	0.0009
	10	0.15±0.053	0.09-0.19	0.031	0.0028	0.12±0.017	0.11-0.14	0.010	0.0003	0.08±0.026	0.06-0.11	0.015	0.0007
	20	0.12±0.021	0.10-0.14	0.012	0.0004	0.03±0.026	0.01-0.06	0.015	0.0007	0.02±0.010	0.01-0.03	0.006	0.0001
	30	0.09±0.001	0.06-0.13	0.021	0.0013	0.02±0.010	0.01-0.03	0.006	0.0001	BDL[1]	-	-	-
Sediments	0	0.14±0.040	0.10-0.18	0.023	0.0016	0.11±0.050	0.07-0.18	0.011	0.0005	0.12±0.016	0.11-0.13	0.001	0.0003
	10	0.12±0.036	0.10-0.16	0.020	0.0012	0.02±0.010	0.01-0.05	0.001	0.0002	0.01±0.004	0.009-0.011	0.001	0.0001
	20	0.03±0.022	0.01-0.06	0.015	0.0007	0.03±0.011	0.02-0.05	0.004	0.0001	0.02±0.009	0.01-0.03	0.011	0.0001
	30	BDL	-	-	-	BDL	-	-	-	BDL	-	-	-
Fish	0	0.11±0.010	0.09-0.15	0.031	0.0013	0.08±0.055	0.07-0.09	0.001	0.0003	0.08±0.050	0.05-0.12	0.001	0.0001
(*Oreochromis*	10	0.04±0.030	0.02-0.07	0.005	0.0013	0.03±0.010	0.01-0.07	0.005	0.0004	BDL	-	-	-
nilotica Lin.)	20	BDL	-	-	-	BDL	-	-	-	BDL	-	-	-
	30	BDL	-	-	-	BDL	-	-	-	BDL	-	-	-
Yams	0	0.30±0.001	0.20-0.50	0.022	0.0020	0.28±0.014	0.26-0.31	0.002	0.0003	0.29±0.003	0.27-0.36	0.001	0.0001
(*Dioscorea*	10	0.24±0.080	0.18-0.30	0.030	0.0012	0.20±0.005	0.17-0.23	0.003	0.0001	0.15±0.010	0.11-0.20	0.017	0.0003
alata)	20	0.12±0.034	0.10-0.14	0.010	0.0015	0.10±0.001	0.06-0.15	0.003	0.0004	0.01±0.001	0.008-0.013	0.005	0.0006
	30	BDL	-	-	-	BDL	-	-	-	BDL	-	-	-

[1]BDL-below detection limit

Table 2(on next page)

Toxicity indices of mercury from consumption of fish and yams from Namukombe stream by adults

1

Table 2. Toxicity indices of mercury from consumption of fish and yams from Namukombe stream by adults

Distance (m)	Fish (*Oreochromis nilotica* Lin.)						Yams (*Dioscorea alata*)					
	EDI (μg/kg/day)			THQ			EDI (μg/kg/day)			THQ		
	Up sluice	Middle sluice	Down Sluice	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down s
0	0.0183	0.0133	0.0133	0.04576	0.03325	0.03325	0.1279	0.1194	0.1237	0.320	0.299	0.30
10	0.0067	0.0049	-	0.01675	0.0123	-	0.1023	0.0853	0.0639	0.256	0.213	0.1
20	-	-	-	-	-	-	0.0512	0.0426	0.0042	0.128	0.105	0.0
30	-	-	-	-	-	-	-	-	-	-	-	-

2

3

4

Table 3(on next page)

Toxicity indices of mercury from consumption of fish and yams from Namukombe stream by children

Table 3. Toxicity indices of mercury from consumption of fish and yams from Namukombe stream by children

Distance (m)	Fish (*Oreochromis nilotica* Lin.)						Yams (*Dioscorea alata*)					
	EDI (μg/kg/day)			THQ			EDI (μg/kg/day)			THQ		
	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down s
0	0.073	0.053	0.053	0.183	0.133	0.133	0.394	0.367	0.381	0.985	0.918	0.9
10	0.027	0.020	-	0.0665	0.05	-	0.315	0.262	0.197	0.788	0.655	0.4
20	-	-	-	-	-	-	0.158	0.131	0.013	0.395	0.328	0.0
30	-	-	-	-	-	-	-	-	-	-	-	-

Table 4(on next page)

Toxicity indices of mercury from dermal contact with dredged sediments in Namukombe stream by adults and children

Table 4. Toxicity indices of mercury from dermal contact with dredged sediments in Namukombe stream by adults and children

Distance (m)	Adults						Children					
	ADD *therm* (μg/kg/day) $\times 10^{-6}$			THQ $\times 10^{-4}$			ADD *therm* (μg/kg/day) $\times 10^{-6}$			THQ $\times 10^{-4}$		
	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down s
0	7.105	5.583	6.090	7.105	5.583	6.090	5.227	4.107	4.480	5.227	4.107	4.48
10	6.090	1.015	0.5075	6.090	1.015	0.5075	4.480	0.747	0.373	4.480	0.747	0.37
20	1.523	1.523	1.015	1.523	1.523	1.015	1.120	1.120	0.747	1.120	1.120	0.74
30	-	-	-	-	-	-	-	-	-	-	-	-

Table 5(on next page)

Bioconcentration factor and Biota to Sediment Accumulation Factor for fish, water and sediment from Namukombe stream

Table 5. Bioconcentration factor and Biota to Sediment Accumulation Factor for fish, water and sediment from Namukombe stream

Distance (m)	Bioconcentration factor			Biota to Sediment Accumulation Factor		
	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down sluice
0	0.091	0.444	0.800	0.786	0.727	0.667
10	0.267	0.250	-	0.333	1.500	-
20	-	-	-	-	-	-
30	-	-	-	-	-	-

Table 6(on next page)

Contamination factor and Muller geoaccumulation index of sediments from Namukombe stream

Table 6. Contamination factor and Müller geoaccumulation index of sediments from Namukombe stream

Distance (m)	Contamination factor			Geoaccumulation index		
	Up sluice	Middle sluice	Down sluice	Up sluice	Middle sluice	Down sluice
0	0.56	0.44	0.48	-1.423	-1.771	-1.644
10	0.48	0.08	0.04	-1.644	-4.230	-5.233
20	0.12	0.12	0.08	-3.644	-3.644	-4.230
30	-	-	-	-	-	-

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